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(19)

(54) MELAMINE RESIN FOAM COMPOSITIONS

(71) We, MELAMINA ULTRA S.A. INDUSTRIA QUIMICA, a Brazilian Body Corporate, of Rua dos Algibebes, 6-sala 304, Caixa Postal, 1616-Salvador, Brazil, and COORDENACAO DOS PROGRAMAS DE POSGRADUACAO DE ENGENHARIA DA UNIVERSIDADE FEDERAL DO RIO DE JANEIRO, —COPPE/UFRJ of Centro de Tecnologia, Bloco G Ilha do Fundao, Caixa Postal 1191, ZC-00 Rio de Janeiro Guanabara, Brazil, do hereby declare the invention, for which we pray that a patent may be granted to us ,and the method by which it is to be performed, to be particularly described in and by the following statement: —

This invention relates to rigid closed cell foams in particular rigid foams based on melamine-form-aldehyde resins, optionally containing various fillers and also to a process for manufacturing such compositions.

According to the present invention there is provided a rigid closed cell foam obtained by the polymerisation of a mixture comprising water, one or more melamine-formaldehyde prepolymers and an acid surface active agent, said foam having a compressive strength of 5.0 to 80 kg/cm², as hereinafter defined.

The mixture from which the rigid foam is obtained may additionally comprise a catalyst, for example an acid catalyst, for promoting the polymerisation reaction of the melamine-formaldehyde prepolymer.

In a further aspect of the present invention there is provided a process for preparing a rigid foam which process comprises expanding a mixture of water, an acid surface active agent and one or more melamine - formaldehyde prepolymers, and allowing the prepolymer to polymerise to form a rigid closed cell foam having a compressive strength of 5.0 to 80 kg/cm², as hereinafter defined.

Preferably the foam comprises one or more inert fillers which may be inorganic or organic fillers.

In this specification, the compressive strength of the foam is defined according to the result obtained by the testing method of the International Union of Testing and Research Laboratories for Materials and Structures (RILEM RECOMMENDATION CPC 4) (1) Materials and Stuctures vol. 6, No. 30, 1972.

In the process for preparing the rigid foam the water and the surface active agent may be initially mixed together and agitated to form a foam. Alternatively the water and surface active agent may be initially mixed together and a gas bubbled into the mixture to form a foam. Furthermore, the water and surface active agent may be initially mixed together and a gas generated in situ in the mixture to form a foam.

A solution of the melamine-formaldehyde prepolymer may then be added to the foam which polymerises to yield a rigid closed cell foam.

Preferably the water and surface active agent are initially mixed together with a volatile organic solvent, a solution of melamime - formaldehyde prepolymer is added to the mixture, the polymerisation reaction of the prepolymer being sufficiently exothermic to expand the reaction mixture.

The foams of the present invention are rigid materials of various densities, based on melamine-formaldehyde resins and optionally various inorganic fillers which have slow or difficult combustion. The fillers may be added to the foam of the melamine-formaldehyde prepolymer and after curing thereof, foams may be obtained, which can be used extensively in the building industry in the form of constructional materials such as partition wals, lowered ceilings and linings, bricks for indoor and outdoor walls, and decora-

tive covering and coatings of decorative character. It has been found that the rigid foams of the present invention have a relatively high mechanical strength and exhibit excellent acoustical and thermal insulating characteristics.

The presently commercially available products for the fulfilment of the above mentioned purposes present various inconveniences, the major one of them being the flammability of the products. The products obtained by the present invention, while under the action of a direct flame have been found to undergo carbonization and were self-extinguishing, i.e., the combustion reaction immediately stopped when the direct flame was withdrawn from the material. Moreover, they possessed the features of nitrogenated materials which when decomposing give rise to the formation of nitrogen and other nitrogenated compounds which retard the oxidation reaction of the material. This reaction was still further hindered by the formation of a charred or carbonized material layer on the surface contacted by the flame and which acted, therefore, as an insulating material against the propagation of the flame. The structural strength of the material did not undergo any noticeable reduction. The inorganic fillers which may be present in the foam may be selected according to the use to which the material is intended and may comprise, for example, gypsum, asbestos, glass and polymeric material fibres, metal powder, magnesite, rockwool, diatomite, rock powder, sand, talc. The organic fillers may be, for example, piassava, cocoanut, and sisal fibres. and begasse of sugar-cane. These fillers may have different agglomeration conditions and fibre length and may be added alone or in appropriate proportions of two or more of these materials.

The present invention will now be described by way of example only, in the following specific example illustrating a process for obtaining a hardenable melamine-formaldehyde prepolymer composition with various fillers.

Example

a) *Preperation of the prepolymer solution*

The melamine-formaldehyde prepolymer, basis of the melamine foam to be produced, was prepared according to the following formulation and operation conditions.

Formaldehyde in 37% (weight/weight) aqueous solution was heated at about 78± 1°C. in a reactor provided with constant agitation and temperature control. Melamine was then added to the formaldehyde neutralized with a 10% (weight/volume) sodium hydroxide aqueous solution and the reactor temperature was raised to 87± 1°C. over a period of about 30 minutes and maintained at this value, the reaction being carried out under reflux. The degree of prepolymerization of the monomer was controlled by turbidity tests (cloud tests) which were frequently carried out during the operation, the details of the test being given below. The end of the reaction was characterized by the turbidity and weight ratio of melamine-formaldehyde prepolymer solution to distilled water of 1:1. The solution was then rapidly cooled to room temperature and the product stored for later use.

The physical characteristics of the prepolymer solution obtained were the following: Clear solution which, in the course of time, became cloudy and hardened, pH = 9.0, density = 1.25 g./cm³, viscosity = 1.2 g./cm/sec both measured at 30±0.1°C, solid contents of about 60% by weight. The molar ratio of the reagents was as follows: 11 moles formaldehyde: 8.63 moles melamine: 0.165 mole sodium hydroxide.

Cloud test

A certain amount of prepolymer solution was weighed and water was slowly added thereto until a premanent cloudiness of the mixture was observed.

The weight ratio of water required to cause this cloudiness or turbidity should be 1:1.

b) *Preparation of rigid melamine resin foam composition*

To a previously measured quantity of water was added an acidic surface active agent which, is this case, was an anionic acid detergent. The mixture was vigorously agitated for about 10 minutes whereupon it acquired the consistency of a light foam. Instead of submitting the mixture to agitation however one may also bubble a gas into the mixture and the same light foam may be obtained. This gas may be supplied from an external source or may be produced *in situ* by means of a chemical reaction or, alternatively by using a low boiling point organic solvent.

The prepolymer solution was then added to the foam obtained above without interrupting the agitation or gas bubbling. Where a volatile organic solvent was used, the polymerization reaction was sufficiently exothermic to cause the blowing i.e. expansion of the reaction mixture. An acid catalyst was then added to the prepolymer solution, however it may be added subsequently in a third step without interrupting the agitation which should last for a sufficient time in order that the reaction mixture be homogenious.

A filler was ultimately added to the foam, care being taken to keep the mixture continuously agitated in order to obtain a homogenous mixture. The composi-

tion may then be moulded into a desired shape.

If pure melamine formaldehyde foam is desired the addition of one or more fillers may be omitted.

Typical proportions or relative amounts of prepolymer, water, surface active agent, acid catalyst and filler, e.g., an inert mineral powder, as used in the present invention are given in the Table below;

Table

Components	*Parts by weight*
Melamine-formaldehyde prepolymer(s)	100
Acidic surface active agent	2 to 10
Water	100 to 800
Acid catalyst	0 to 2.5
Filler	0 to 200

In general the properties of a rigid foam composition prepared as described above may be listed as follows:

a) The colour of the material usually depends upon the filler added. In the case where the foam has no filler added thereto the colour is generally white. The product has been found to exhibit low density, the fragility of the material increasing with a decrease of its density, but even in formulations of low density the material may withstand manual compression and tension.

b) The product has been found to withstand the action of water and corrosive agents and it does not undergo modifications in its properties when in prolonged contact with such agents.

c) The apparent density of the material is generally in the range of 0.05 g./cm^3 up to densities of about 1.00 g./cm^3, depending upon the quantity of water and surface active agent of the formulation.

d) The product had a compressive strength of 5.0 kg/cm^2 to 80 kg/cm^2, increasing linearly with the density.

e) The material had an internal cellular structure, formed of closed cells, which gives it exceptional thermal and acoustical properties.

Its external surface, however, showed an attractive appearance with a bright coat of resin without discontinuities.

f) The material was not combustible.

g) The surface of the material may be painted by conventional methods and products or even coated with commercially available materials such as decorative wall-paper, metal foils and wooden sheets and even other types of finishing with laminated plastic material.

WHAT WE CLAIM IS:—

1. A rigid closed cell foam obtained by the polymerisation of a mixture comprising water, one or more melamine-formaldehyde prepolymers and an acid surface active agent, said foam having a compressive strength of 5.0 to 80 kg/cm^2 as hereinbefore defined.

2. A foam as claimed in claim 1 which additionally comprises an inert filler.

3. A foam as claimed in claim 2 wherein the inert filler is an inorganic filler.

4. A foam as claimed in claim 2 wherein the inert filler is an organic filler.

5. A foam as claimed in any one of the preceding claims and obtained by the polymerisation of the mixture additionally comprising a catalyst for promoting the polymerisation reaction of the melamine-formaldehyde prepolymer.

6. A foam as claimed in claim 5 wherein the catalyst is an acid catalyst.

7. A foam as claimed in claim 5, the mixture comprising for each 100 parts by weight of melamine-formaldehyde prepolymers, from 2 to 10 parts by weight of the surface active agent, from 100 to 800 parts by weight of water, from 2 to 5 parts by weight of an acid catalyst for promoting the polymerisation reaction and up to 200 parts by weight of an inert filler.

8. A process for preparing a rigid foam which process comprises expanding a mixture of water, an acid surface active agent and one or more melamine-formaldehyde prepolymers, and allowing the prepolymer to polymerise to form a rigid closed cell foam having a compressive strength of 5.0 to 80 kg/cm^2, as herinbefore defined.

9. A process as claimed in claim 8 wherein the water and the surface active agent are initially mixed together and agitated to form a foam.

10. A process as claimed in claim 8 wherein the water and surface active agent are initially mixed together and a gas is bubbled into the mixture to form a foam.

11. A process as claimed in claim 8 wherein the water and surface active agent are initially mixed together and a gas is generated in situ in the mixture to form a foam.

12. A process as claimed in any one of claims 9 to 11 wherein a solution of the melamine-formaldehyde prepolymer is added to the foam which polymerises to yield a rigid closed cell foam.

13. A process as claimed in claim 8 wherein the water and surface active agent are initially mixed together with a volatile organic solvent, a solution of melamine-formaldehyde prepolymer is added to the mixture, the polymerisation reaction of the prepolymer being sufficiently exothermic to expand the reaction mixture.

14. A process as claimed in any one of claims 8 to 13 wherein a catalyst for promoting the polymerisation reaction is

added to the mixture of water, surface active agent and melamine-formaldehyde prepolymer.

15. A process as claimed in any one of claims 8 to 14 wherein one or more inert fillers is added to the mixture before polymerisation of the prepolymer is complete.

16. A process for preparing a rigid foam as claimed in claims 8 substantially as hereinbefore described in the specific example.

17. A rigid foam as claimed in claim 1 substantially as herein before described in the specific example.

18. A rigid foam whenever prepared by a process as claimed in any one of claims 8 to 16.

19. Shaped or moulded articles comprising a rigid foam as claimed in any one of claims 1 to 7 or 17 and 18.

20. Constructional materials whenever comprising a rigid foam as claimed in any one of claims 1 to 7 or 17 and 18.

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